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AEROSOLS AND CLOUDS IN CHEMICAL TRANSPORT MODELS AND CLIMATE MODELS

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Aerosols and Clouds in Chemical Transport Models and Climate Models

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Abstract

Clouds exert major influences on both shortwave and longwave radiation as well as on the hydrological cycle. Accurate representation of clouds in climate models is a major unsolved problem because of high sensitivity of radiation and hydrology to cloud properties and processes, incomplete understanding of these processes, and the wide range of length scales over which these processes occur. Small changes in the amount, altitude, physical thickness, and/or microphysical properties of clouds due to human influences can exert changes in Earth's radiation budget that are comparable to the radiative forcing by anthropogenic greenhouse gases, thus either partly offsetting or enhancing the warming due to these gases. Because clouds form on aerosol particles, changes in the amount and/or composition of aerosols affect clouds in a variety of ways. The forcing of the radiation balance due to aerosol-cloud interactions (indirect aerosol effect) has large uncertainties because a variety of important processes are not well understood precluding their accurate representation in models.

1. Introduction

Clouds are an extremely important element of Earth's climate system. Clouds are highly reflective in the solar spectrum, yet strongly absorbing in the thermal infrared; consequently, they produce a large impact on Earth's radiation budget. This impact is called cloud radiative forcing (CRF). Satellite studies have shown that, on global average, clouds decrease absorption of solar radiation by about 50 W m^{-2} (shortwave CRF) and decrease upwelling thermal infrared radiation by 30 W m^{-2} (longwave CRF), thus exerting net CRF of about –20 W m^{-2} (Kiehl and Trenberth, 1997). Locally and instantaneously, clouds can reduce absorbed shortwave radiation by as much as 700 W m^{-2}. Clouds also play a central role in Earth's hydrological cycle, which is coupled to the energy budget through latent heat of water condensation/evaporation, influencing circulations on a variety of scales. The nature and extent of these cloud processes may be expected to change in the future in response to changes in concentrations and properties of trace gases and aerosols and resulting changes in climate. For these and other reasons, accurately representing clouds and their radiative and hydrological influences in climate models is essential. However for a variety of reasons accurate representation of clouds and cloud influences on radiation and hydrology in climate models remains particularly challenging. Key among these reasons are the fact that the condensed phase (liquid, solid) of clouds is a small fraction of total water present in the cloud, necessitating accurate representation of both total water content and temperature, governing saturation vapor concentration; complexities associated with the presence of several phases of condensed-phase water (liquid, ice, mixed); the spatial and temporal diversity of cloud microphysical structure, as reflected in the number concentration and size distribution of cloud hydrometeors and the crystal habit of ice clouds; and the numerous varieties and morphologies of clouds and the resultant complexity of their three-dimensional structure on many scales, Figure 1.

Figure 1. Complexity of three-dimensional structure of clouds; note penetration of cumulonimbus clouds through thin cirrus layer. Photo courtesy of Y.-N. Lee, Brookhaven National Laboratory.

Small changes to macrophysical properties (coverage, structure, altitude) and microphysical properties (droplet size, phase) can exert substantial effects on climate. For instance a 5% increase of the shortwave cloud forcing, as might occur as a consequence of changes in the nature and amount of atmospheric aerosols, would compensate the increase in greenhouse gases between the years 1750–2000 (Ramaswamy et al., 2001). Recognition of this situation has stimulated the development of improved physically based representations of cloud processes generally and of aerosol influences on clouds for inclusion in climate models. However, despite intensified research, the lack of understanding feedbacks of external forcings on clouds remains one of the largest uncertainties in climate modeling projections of future and climate change (Randall et al., 2007). Likewise understanding the radiative forcing by aerosols through their influences on clouds remains the greatest uncertainty in radiative forcing of climate change over the industrial period (IPCC, SPM 2007).

The principal tools for examining prospective consequences of future emissions of greenhouse gases and aerosols on Earth's climate are general circulation models (GCMs); the acronym GCM also often stands for global climate model, and the terms are often used interchangeably. These models are not only the primary tool for simulation of global climate change, but they are also important for evaluating the regional effects of anthropogenic emissions on modifying precipitation distribution and amounts. By integrating atmospheric, radiative, oceanic, and land-surface processes on the global scale they provide an indication of expected changes in the coupled system, including possible consequences of coupled increases in greenhouse gases and aerosols on atmospheric radiation, on clouds and precipitation and on the climate system generally. This paper examines the present state of understanding of aerosol and cloud processes that must be represented in GCMs and the state of such representation and identifies key recent advances and needed developments.

The first requirement of using GCMs to examine aerosol influences on clouds and precipitation is that the models accurately represent the macrophysical properties of clouds and precipitation, including their geographical and seasonal variation. Although GCMs have been used to examine the influence on global climate of widespread anthropogenic sources of cloud condensation nuclei, CCN, the aerosol particles that serve as the nuclei on which cloud

droplets form, such examination presents numerous problems. First is the issue of scales. GCMs grid cells typically have horizontal dimension of 150 km to 250 km and vertical dimension of hundreds to thousands of meters, over which there can be substantial spatial inhomogeneity. For example clouds often cover a small fraction of the volume of a grid cell necessitating rather ad hoc parameterizations, and the average vertical velocities in a grid cell are very small (~0.01 m s^{-1}) whereas actual vertical velocities, which control cloud formation and the activation of aerosol particles to cloud droplets, might be 1 m s^{-1} or greater. The poor representation of convection is likely a major source of error in modeled liquid and solid water in clouds.

It is clear (Figure 2) that there are major disparities amongst GCMs, even in zonal averages of cloud albedo, which is a major determinant of Earth's radiation budget. Here each panel corresponds to a different GCM; The model output was obtained from coordinated simulations with 20 different coupled ocean-atmosphere GCMs, performed in support of the IPCC Fourth Assessment Report. Clearly these models cannot all be correct. Although space-based measurements can identify models that are doing better or worse relative to this important cloud variable, such measurements are also difficult, although uncertainties in observations are smaller than the inter-model differences.

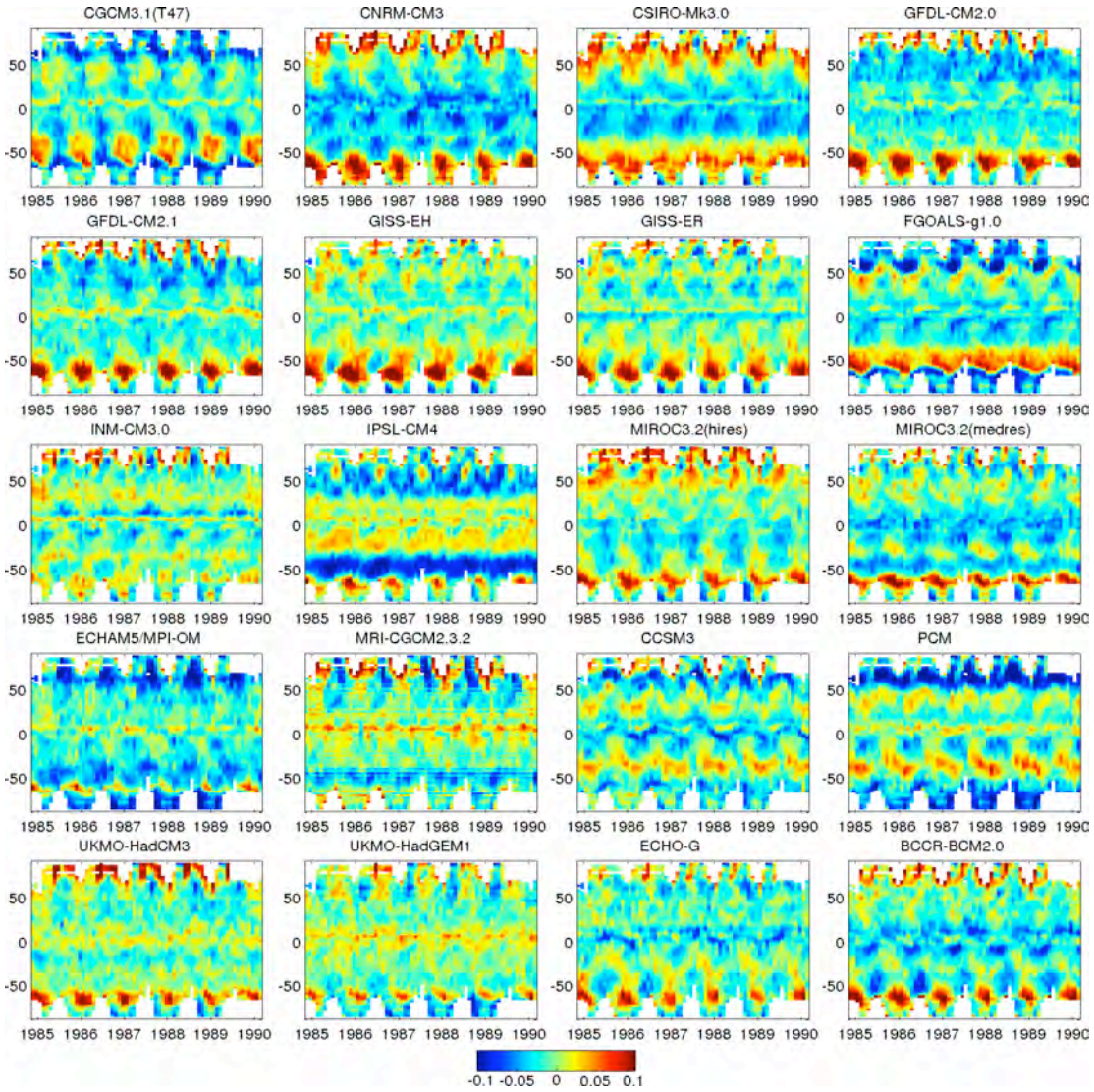

Figure 2. Difference between cloud albedo as determined by satellite measurements (Earth Radiation Budget Experiment, ERBE) and twenty global climate models as a function of latitude and time (November 1984 –February 1990). Positive anomalies, where ERBE is higher, are indicated with red, and negative anomalies, where ERBE is lower, with blue colors. Courtesy of Frida Bender. Modified from Bender at al, 2006.

Cloud microphysical properties are determined by processes such as droplet and crystal nucleation, condensation, evaporation, gravitational settling, and precipitation, all of which operate at the scale of the individual cloud particles or of local populations (less than one meter). In contrast, the spatial distribution of clouds is determined by dynamical processes such as turbulence, updrafts, downdrafts, and frontal circulations and by radiative cooling, which operate across scales from meter to global. However these scales are coupled by a variety of processes, such as microphysical influences on precipitation development, that in turn affect latent heat release below cloud, that again affects atmospheric stability and vertical motions. The treatment of these processes in climate models and the confidence in this treatment are limited by both lack of understanding and computational resources to represent these processes at all relevant scales, the latter necessitating development and application of parameterizations, which are inherently scale dependent. The requirement of accurately representing the many roles of clouds in the climate system and more generally in the

biogeochemistry of the planet attaches not only to the present atmosphere but also to prior atmospheres (necessary for evaluation of performance of climate models over the instrumental record of the past 150 years or so) and to future atmospheres (necessary to evaluate the influences of different projected emissions scenarios of greenhouse gases and aerosols). A concern is that each role's common dependence on many of the same cloud properties and processes suggests that errors in simulating one role would produce errors in other roles. Conversely, improving cloud treatment to reduce uncertainty in one role also will reduce uncertainty in other roles. Hence, improving representations and parameterizations of cloud processes in climate models will produce many benefits well beyond the simulation of cloud feedbacks and of aerosol indirect effects.

2. Representation of aerosols in global-scale chemical transport models (CTMs) and global climate models (GCMs)

Although there are many similarities between treatment of aerosol processes in CTMs and GCMs, it is useful to distinguish the two modeling approaches. GCMs simulate their own meteorology and couple aerosol cycles with clouds, precipitation and the radiation budget. Their strength therefore lies in the projection of future climate for different emissions scenarios. Because the emphasis of GCMs is on the long-term simulation of climate, treatment of aerosol processes in climate models must be greatly simplified. In contrast, with CTMs it is possible to treat aerosol processes and interactions between aerosols (and hydrometeors) and atmospheric chemistry in greater detail. CTMs are often driven by observed meteorology; in such models the aerosol chemistry and physics do not feed back on the meteorology. CTMs and GCMs need to be driven by observed meteorology in order to capture detailed aerosol processes and to compare simulated aerosol fields with observations.

Since the pioneering study by Langner and Rodhe (1991), who used a coarse horizontal resolution CTM based on climatological meteorology to represent the global distribution of the mass concentration of sulfate aerosol (without explicit representation of aerosol microphysics), there have been substantial advances in the complexity of the treatment of many key processes: aerosol precursor chemistry, aerosol microphysical processes, transport processes, and particle dry deposition and wet removal. Recently attempts have been undertaken to calculate not just the aerosol mass concentration but also the particle number concentration by parameterizing aerosol formation and dynamical processes. An overview of the processes with most be understood and represented in models is given in Figure 3.

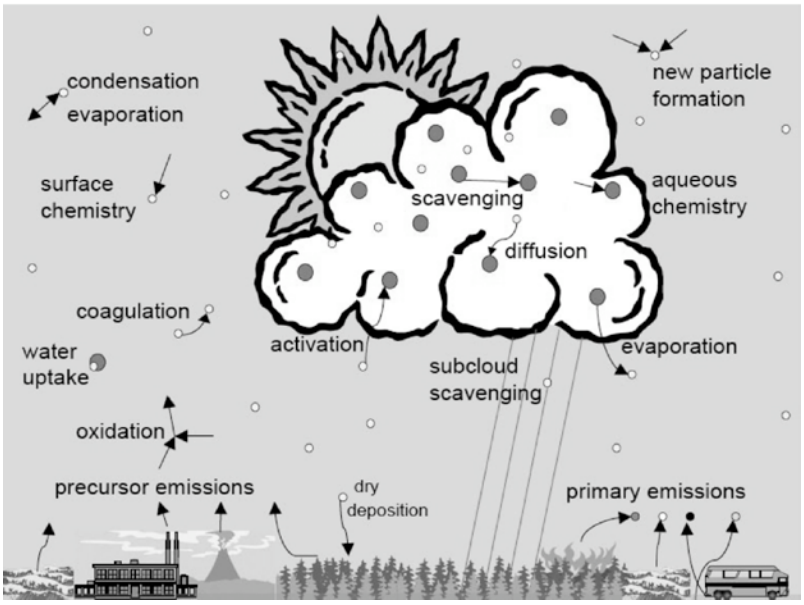

Figure 3: Important climate-influencing aerosol processes that must be accurately represented in climate models. Aerosol particles are directly emitted as primary particles and are formed secondarily by oxidation of emitted gaseous precursors. The formation of low-volatility materials results in new particle formation and condensation onto existing particles. Aqueous-phase oxidation of gas-phase precursors within cloud droplets accretes additional mass onto existing particles but does not form new particles. Particles age by surface chemistry and coagulation as well as by condensation. The uptake of water with increasing relative humidity increases particle size, affecting also the particle optical properties. As cloud form some fraction of aerosol particles are "activated" to form cloud droplets. Within clouds interstitial particles can become attached to cloud droplets by diffusion, and activated particles are combined when cloud droplets collide and coalesce. If cloud droplets evaporate the particles remain in the atmosphere; if the cloud precipitates the particles are carried below the cloud and reach the surface, unless the precipitating particles completely evaporate. Aerosol particles below precipitating clouds can also be removed from the atmosphere by impaction by precipitating drops and by dry deposition to the surface (from Ghan and Schwartz, 2007).

Most of the earlier studies concerned with the effect of aerosol particles on the climate system have just taken sulfate particles into account or have considered sulfate as a surrogate for all anthropogenic aerosols. Lately most major GCMs include also carbonaceous aerosols, dust and sea salt (for a synopsis of the state of model development see: the AEROCOM model intercomparison project: http://nansen.ipsl.jussieu.fr/AEROCOM/ and Kinne et al., 2006). This project has enabled comparison of the results of aerosol simulations from more than a dozen modeling groups world wide. An example of such a comparison is given in Figure 4 for global and annual mean aerosol optical depth, the vertical integral of aerosol extinction coefficient. Although the figure indicates fairly good agreement with observations for most of the models, it is clear that there are substantial differences in the contributions of the several aerosol species.

A major source of uncertainty in present aerosol modeling is the lack of accurate time-resolved emission inventories. In particular, biogenic sources and emissions from biomass burning are highly uncertain. Both biogenic and biomass burning emissions depend on environmental conditions (e.g. weather) and exhibit high interannual variability, which has not been taken into account by climate studies. Probably the largest uncertainty is associated with organic aerosols because current measurement techniques cannot identify the many

organic species present in primary emissions (Kanakidou et al., 2004). A second issue is that the chemical pathways in the atmosphere are complex and not fully understood. Organic aerosols result both from primary emission and from gas-to particle conversion in the atmosphere (secondary production). The total source of these organic aerosols is thus a major wildcard in simulations of future scenarios. Advances in measurement techniques for aerosols are thus of critical importance. One recent such advance is the aerosol mass spectrometer, which is permitting development of a systematic measurement data base of the composition of aerosols generally and identification of primary and secondary organic species (Zhang et al., 2007). Simulating nitrate aerosols remains problematic because of their semi-volatile nature. In addition to all the difficulties in developing understanding of the chemical and microphysical processes, simulation of aerosol processes in large-scale models is highly CPU-time consuming.

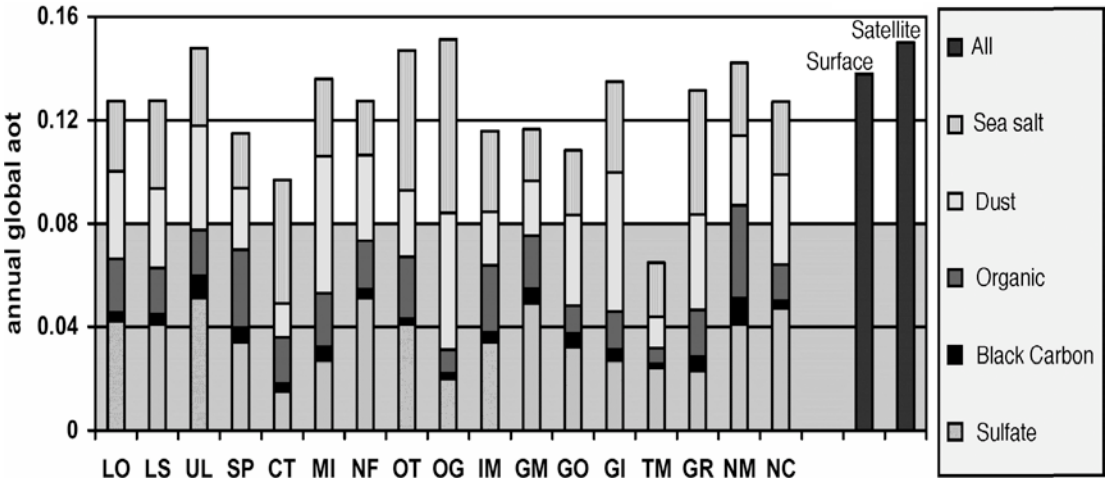

Fig. 4. Simulated contributions of five aerosol components (seasalt, dust, organic, black carbon, and sulfate) to annual and global mean aerosol optical thickness (at 550 nm) by 17 CTMs. Shown also for comparison are surface measurements by the AERONET network and a composite of satellite measurements. Modified from Kinne et al., 2006, which gives citations to the individual model studies.

There is increasing evidence that individual aerosol particles consist predominantly of a conglomerate of multiple internally mixed chemical substances. In contrast most GCMs still treat aerosols as external mixtures because internal mixtures have more degrees of freedom, are more complex, and impose an added computational burden. however the mixing state of aerosol particles (externally versus internally mixed) strongly influences their optical properties and their ability to act as CCN. For example, a slight coating of a particle by an only moderate soluble organic species can drastically increase its ability to act as a CCN. Therefore, treating the degree of mixing properly is essential for accurately representing aerosol processing in GCMs, including aerosol-cloud interactions. Advanced aerosol modules in some GCMs have been expanded to include aerosol mixtures (see Lohmann and Feichter, 2005 for references).

Representing the size distributions of aerosols and their evolution is also essential. Two kinds of aerosol dynamics models have been developed: modal schemes and bin schemes. Modal schemes represent each aerosol mode with a log-normal distribution of aerosol mass and possibly number. Bin schemes divide the aerosol spectrum in a large number of bins. Modal representations of the aerosol size distribution typically evolve aerosol number concentration in addition to mass concentration.

3. Representation of cloud microphysical processes in global climate models (GCMs)

3.1 Microphysics of large-scale clouds

Major improvements have recently been made in the description of cloud microphysics for large-scale clouds. Whereas early studies diagnosed cloud amount based on relative humidity, most GCMs now explicitly calculate cloud condensate in large-scale clouds. The degree of sophistication varies from calculating the sum of cloud water and ice to calculating cloud water, cloud ice, snow and rain as separate species (see Lohmann and Feichter, 2005 for references). Because the aerosol indirect effect is based on the change in cloud droplet number concentration, some GCMs explicitly calculate cloud droplet number concentrations in addition to the cloud water mass mixing ratio using one of the above described physically based aerosol activation schemes as a source term for cloud droplets. Likewise the number of ice crystals needs to be calculated in addition to the ice water mass mixing ratio in order to estimate the effect of aerosols on mixed-phase and ice clouds. Determining the size-dependent sedimentation rate of hydrometeors requires at least a two-moment scheme; representing this size-dependent sedimentation leads to important differences in the cloud vertical structure, cloud lifetime and cloud optical properties. Two-moment schemes are superior to one-moment schemes provided that the second moment can be treated adequately. However, there are still major uncertainties regarding activation of cloud droplets and precipitation formation, as discussed below. Theoretically, the best approach would be to use a size-resolved treatment of the cloud microphysics, but taking this approach in a GCM would be questionable because treatment of cloud dynamics, including entrainment and advection, is not accurate enough to warrant such a level of detail.

3.2 Microphysics of convective clouds

There is currently a substantial discrepancy between the degree of sophistication in cloud microphysics in large-scale clouds and the very rudimentary treatment of cloud microphysics in convective clouds, perhaps because stratiform clouds are generally much more susceptible to indirect effects than are convective clouds. However recently evidence has emerged that biomass burning affects convective clouds, necessitating improvements in the treatment of microphysical processes in convective clouds. A first global study (Nober et al. 2003) accounted for this effect by decreasing the precipitation efficiency for warm cloud formation in convective clouds depending on the cloud droplet number concentration. This approach was taken a step further by Lohmann (2007) who introduced the same microphysical processes (nucleation, autoconversion, freezing, aggregation, etc) that are considered in large-scale clouds into convective clouds as well.

Another option, though considerably more computationally expensive, is to use so-called "super-parameterizations", in which cloud resolving models are embedded within the normal GCM grid cells, but at only a small fraction of the area of the parent GCM grid cell (e.g. Randall et al. 2003). These models have the capability of calculating cloud-scale vertical velocities and LWC and thus explicitly representing precipitation processes. However they have yet to be applied to aerosol effects on precipitation. If the representation of aerosol and clouds in such models, or others using new and innovative techniques for representing subgrid processes can be improved, they should increase the accuracy of calculations of the influence of aerosols on the amount and distribution of clouds and precipitation as well as on radiation.

3.3 Cloud droplet formation

Linking aerosol particles to cloud droplets is a weak point in estimates of the indirect aerosol effects. Accurate treatment of cloud droplet formation requires knowledge of the particle number concentration and size distributed chemical composition of the aerosol and the vertical velocity on the cloud scale. Parameterizations based on Köhler theory have been

developed that can describe cloud droplet formation for a multi-modal aerosol. This approach has been extended to include kinetic effects that take into account mass accommodation of water at the gas-liquid interface and also account for the fact that the largest particles do not have time to grow to their equilibrium size and activate. Also, the competition between natural and anthropogenic aerosols, such as between sulfate and sea salt, is considered (Forster et al., 2007).

Organic carbon is an important constituent of cloud condensation nuclei; especially if it is surface active. Facchini et al. (1999) indicated that a lowering of the surface tension of some surface-active organic aerosols as obtained from fog water samples would enhance the cloud droplet number concentration and cloud albedo and hence could lead to a negative forcing as great as ~-1 W m^{-2}. On the other hand, amphiphilic film-forming compounds may retard cloud droplet formation (Feingold and Chuang, 2002). The delayed activation enables the growth of a mode of larger drops that formed earlier on and therefore leads to increased dispersion and enhanced drizzle formation. Chemical effects on cloud droplet formation and thus on the indirect effect may be as large as the effects of unresolved cloud dynamics (see Lohmann and Feichter, 2005 for references). While the effect of surface active organics has recently been included in parameterizations of cloud droplet formation (Abdul-Razzak and Ghan, 2004), other effects of organics, such as their film-forming ability have not yet been treated.

Application of parameterizations of cloud-drop activation requires estimation of cloud-scale vertical velocities in models that do not resolve these cloud scales. In recognition that this information may not be available some modelers have assumed an empirical relationship between modeled sulfate mass concentrations and droplet concentrations (e.g. Boucher and Lohmann 1995), which is equivalent to assuming there is only a single-value of cloud updraft velocity for all clouds in the model. Others have estimated vertical velocity based on turbulent kinetic energy calculated in boundary layer models (e.g. Lohmann et al. 1999). This is a step in the right direction, but it does not take into account the fact that cloudy updrafts are at the tail of the probability density function (PDF) of vertical velocity. Ghan et al. (1997) among others assumed a normal distribution of vertical velocity with a mean value given by the GCM grid point mean. They then determined the velocity-weighted mean droplet concentration that takes into account the tails of their assumed PDF of vertical velocity. However, observed PDFs of vertical velocity in clouds in the boundary layer are multimodal and are better represented by double-Gaussian PDFs (Larson et al. 2001) with a mean that is a function of the root mean square vertical velocity rather than by a GCM grid point mean (Peng et al. 2005).

3.4 Precipitation formation in warm clouds

The influences of precipitation and drizzle processes on cloud lifetime, cloud water content, and cloud radiative properties discussed above cannot be simulated well in current GCM cloud parameterization schemes. For example, precipitation processes are non-linear functions of total condensate water contents. As a result, the mean liquid water content (LWC) from a GCM model grid-box is essentially meaningless for the representation of precipitation production (e.g. Pincus and Klein 2000). As the autoconversion bias due to horizontal heterogeneity has been found to scale strongly with cloud fractional coverage (Wood et al., 2002), it may be overcome using a parameterization that takes this bias into account. On the other hand, a PDF approach to subgrid modeling may be the optimum approach to resolving these deficiencies. PDFs of subgrid quantities, such as vertical velocity and LWP, are determined from prescribed basis functions in which various moments of the basis functions are calculated in the models (e.g. Pincus and Klein 2000).

Autoconversion of cloud water to precipitation is a key process governing the amount and lifetime of clouds in the atmosphere that must be accurately represented in models, from

cloud-resolving scale to global scale. Even though the mass transfer rate of cloud to rain is dominated by accretion in most clouds (Wood, 2005), autoconversion if the dominant process in most GCMs because rain is assumed to reach the surface within one model time step. Thus, developing parameterizations for autoconversion suitable for incorporation in large-scale models is an active area of research. Traditional parameterizations are either empirically or intuitively obtained (e.g., Kessler 1969 and Sundqvist 1978) or are derived by curve-fitting detailed microphysical models with simple functions such as a power-law (e.g., Berry 1968; Beheng 1994). However these parameterizations lack clear physics and have arbitrarily tunable parameters. Furthermore, parameterizations that calculate at least the cloud droplet number concentration in addition to the liquid water content would be expected to provide much better representation of cloud radiative influences and of aerosol effects than existing one-moment schemes.

One promising scheme that has been derived from theoretical considerations (Liu et al., 2007, and earlier papers referenced therein) represents the autoconversion of any integral moment quantity Y of a cloud drop size distribution as

$$P_Y = P_{Y0} T_Y, \tag{1}$$

where P_Y / Y is the fraction of the quantity Y that is autoconverted per second, P_{Y0} is the rate function describing the conversion rate after the onset of the autoconversion process, and T, ($0 \le T \le 1$), is a threshold function describing the onset of autoconversion. For example, (Figure 5a), the onset of autoconversion of cloud water mass occurs when the mean mass of droplets in the cloud $\overline{m}$ (g) increases beyond a critical value m_c defined as

$$m_c = \frac{m_1}{\kappa^{1/2}} \beta_{con}^{1/2} N^{1/2} L^{-1}. \tag{2}$$

In this expression m_1 is the mass of a single water molecule, 3.0×10^{-23} g; L is the cloud liquid water volume fraction (dimensionless); and N is the cloud droplet concentration (cm^{-3}); $\kappa \approx 1 \times 10^{10}$ cm^{-3} s^{-1} is from a parameterization of the rate of decrease in concentration of falling cloud drops of volume x (cm^3), $n(x)$ due to collision and coalescence with smaller cloud droplets of volume $y < x$ having concentration $N(y < x)$ (cm^{-3}), $\partial \ln n(x) / \partial t = -\kappa x^2 N(y < x)$ (Long, 1974); and $\beta_{con} \approx 1$ x 10^{23} s^{-1} is an effective diffusion coefficient representing the Brownian-like motion along the coordinate of droplet size that is due to turbulent fluctuations in the growth-evaporation of the drop (McGraw and Liu, 2006). For typical values of L (1×10^{-7}) and N (100 cm^{-3}) the value of m_c, ~1×10^{-8} g, corresponds to drop radius ~13 μm. The increase in the threshold function with increasing mean droplet mass $\overline{m}$ is due to the onset collision coalescence resulting from differential gravitational settling. The critical droplet mass m_c characterizes the droplet size for which the rate of growth (by both condensation and collection) equals the rate of evaporation. The onset of autoconversion is not abrupt at m_c, as in the Kessler-type parameterization, but occurs over a range of values of $\overline{m}$, depending on the relative dispersion of the cloud drop size distribution (ε, ratio of standard deviation to mean radius). This dependence captures initiation of the autoconversion by large drops at the high end of the size distribution, and its variation encompasses prior empirical representations of threshold behavior. The rate function P_{Y0} needed to calculate the autoconversion rate P_Y shown in Figure 5b, obtained by integrating the continuous collection equation over all the cloud droplets (Liu and Daum, 2004), is likewise a function of L, N, and ε. The parameterization has found application in modeling on regional (Gustafson et al., 2007) and global scales (Rotstayn and Liu, 2005), modeling scavenging of soluble gases by precipitation (Garrett et al., 2006), and remote sensing of precipitation (Berg et al., 2006).

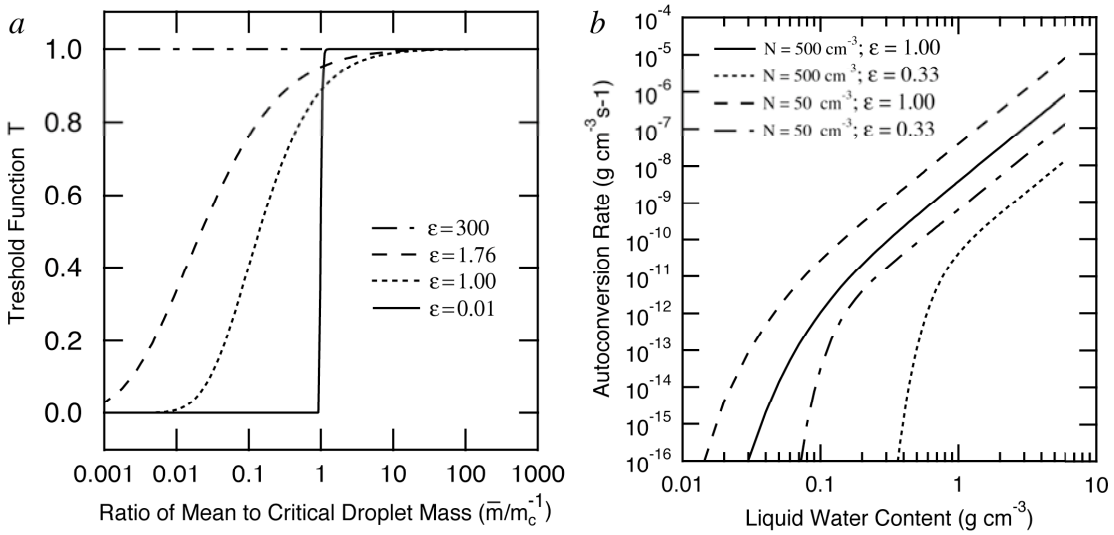

Figure 5. Threshold function (left) and autoconversion rate (right) for cloudwater mass concentration to precipitation; $\overline{m}$ denotes the mean mass of droplets in the cloud, and m_c denotes the critical droplet mass as described in the text; ε denotes relative dispersion of cloud drop size distribution (ratio of standard deviation to mean radius); N denotes cloud drop number concentration. Y. Liu, Brookhaven National Laboratory, private communication, 2008.

4. Aerosol indirect effects

Aerosols affect radiative fluxes by scattering and absorbing solar radiation (direct effect). They also interact with clouds and the hydrological cycle by acting as cloud condensation nuclei (CCN) and ice nuclei. For a given cloud liquid water content, a greater concentration of CCN increases cloud albedo (indirect cloud albedo effect) and is supposed to reduce the precipitation efficiency (indirect cloud lifetime effect), both of which are likely to result in a reduction of the global, annual mean net radiation at the top of the atmosphere. However, these effects may be partly offset by evaporation of cloud droplets due to absorbing aerosols (semi-direct effect) and/or by more ice nuclei (glaciation effect). The influences of these processes on radiation at the top of the atmosphere (TOA) and at the surface and on precipitation are summarized in Table 1. The discussion below is based on Denman et al, (2007), which also provides references to the studies noted.

Table 1. Overview of the several aerosol indirect effects on net radiative flux at the top-of-the atmosphere (TOA) and the surface, and on precipitation, and assessment of level of current scientific understanding. Modified from Denman et al., 2007

Effect	Cloud albedo effect	Cloud lifetime effect	Semi-direct effect	Glaciation indirect effect	Thermodynamic effect
Cloud Types Affected	All; greatest for clouds of intermediate optical thickness	All	All	Mixed-phase	Mixed-phase
Process	For same cloud water or ice content more but smaller cloud particles reflect more solar radiation	Smaller cloud particles decrease precipitation efficiency prolonging cloud lifetime	Absorption of solar radiation by absorbing aerosols evaporates cloud particles, increases static stability,	Increase in ice nuclei increases precipitation efficiency	Smaller cloud droplets delay freezing causing supercooled clouds to extend to colder temperatures
Change in net TOA irradiation	–	–	+ or –	+	+ or –
Potential Magnitude	medium	medium	small	medium	medium
Scientific Understanding	low	very low	very low	very low	very low
Change in surface irradiation	–	–	+ or –	+	+ or –
Potential Magnitude	medium	medium	large	medium	medium
Scientific Understanding	low	very low	very low	very low	very low
Change in Precipitation	n/a	–	–	+	+ or –
Potential Magnitude	n/a	small	large	medium	medium
Scientific Understanding	n/a	very low	very low	very low	very low

Another aerosol influence on clouds and radiation that may be climatologically important is the enhancement of downwelling longwave radiation from thin Arctic stratus whose longwave optical thickness is increased by increased droplet concentration (Lubin and Vogelmann, 2006).

In addition to increasing the number concentration of aerosol particles there is evidence (Liu and Daum, 2002) that increased concentrations of aerosols can increase the breadth of the cloud drop size distribution; this would have the effect of decreasing aerosol influences on shortwave radiation and on inhibition of precipitation development (e.g., Peng and Lohmann, 2003).

The increase in albedo of liquid water clouds due to anthropogenic aerosols has been received much attention. While uncertainties remain regarding the breadth of the cloud drop size distribution, more and probably larger uncertainties are related to aerosol effects on precipitation and on mixed- and ice-phase clouds, as discussed below.

4.1 Aerosol Effects on Water Clouds and Warm Precipitation

Aerosols are hypothesized to increase the lifetimes of clouds because increased concentrations of smaller droplets lead to decreased drizzle production and reduced precipitation efficiency (Albrecht, 1989). It is difficult to devise observational studies that can

separate the cloud lifetime from the cloud albedo effect. Thus, observational studies usually provide estimates of the combined effects. Similarly, climate models cannot easily separate the cloud lifetime indirect effect once the aerosol scheme is fully coupled to a cloud microphysics scheme but likewise calculate the combined cloud albedo, lifetime and semi-direct effect.

GCM studies suggest that in the absence of giant CCN and aerosol-induced changes in ice microphysics anthropogenic aerosols suppress precipitation; precipitation would be suppressed as well as in mixed-phase clouds in which the ice phase plays only a minor role. A reduction in precipitation formation leads also to increased cloud processing of aerosols. Studies with cloud resolving models, on the other hand have shown that cloud processing could lead either to an increase or decrease in precipitation formation in subsequent cloud cycles, depending on the size and concentration of activated CCN. When the actual cloud lifetime is analyzed in cloud resolving model simulations, an increase in aerosol concentration from very clean to strongly anthropogenically influenced situations does not increase cloud lifetime, even though precipitation is suppressed (Jiang et al., 2006). This effect is due to competition between precipitation suppression and enhanced evaporation of the more numerous smaller cloud droplets at high cloud droplet concentration. Giant sea salt nuclei, on the other hand, may override the precipitation suppression effect of the large number of small CCN.

4.2 Aerosol Impacts on Mixed-Phase Clouds

GCM studies suggest that if, in addition to mineral dust, hydrophilic black carbon aerosols are assumed to act as ice nuclei at temperatures between 0 and –35°C, then increases in aerosol concentration from pre-industrial to present times may have resulted in increased glaciation of supercooled stratiform clouds and increase the amount of precipitation via the ice phase. This process could decrease the global mean cloud cover, leading to increased absorption of solar radiation. Whether the glaciation effect or the warm cloud lifetime effect is larger depends on the chemical nature of the dust (Lohmann and Diehl, 2006).

Simulations of precipitation from single cell mixed-phase convective clouds suggest a reduction for various background aerosol concentrations when aerosol concentrations are increased. Khain et al. (2005) postulated that smaller cloud droplets, such as those originating from human activity, would change the thermodynamics of convective clouds. More but smaller droplets would reduce the production of rain in convective clouds. When these droplets freeze, the associated latent heat release would then result in more vigorous convection and more precipitation. In a clean cloud, on the other hand, rain would have depleted the cloud so that less latent heat is released when the cloud glaciates resulting in less vigorous convection and less precipitation. For a thunderstorm in Florida in the presence of Saharan dust, the simulated precipitation enhancement only lasted two hours after which precipitation decreased as compared with clean conditions. This highlights the complexity of the system and indicates that the sign of the global change in precipitation due to aerosols is not yet known. Note that microphysical processes can only change the temporal and spatial distribution of precipitation while the total amount of precipitation can only change if evaporation from the surface changes.

4.3 Subgrid-scale variability and radiative transfer

Model inaccuracy can result from treating clouds as plane parallel homogeneous clouds, as would be the case when clouds are uniformly distributed over grid cells, as is conventional. This treatment can overestimate the Twomey effect by up to 50% (see Lohmann and Feichter, 2005 for references). One way to obviate this problem is to treat grid cells as non uniform. Also here a PDF based approach can be used to account for subgrid-scale variability in cloud

cover and cloud condensate in radiative transfer through inhomogeneous cloud fields (e.g., Pincus and Klein, 2000).

4.4 Aerosol Impacts on Cirrus Clouds

The influence of aerosols from aircraft emissions on cirrus cloud extent and properties has received considerable attention because these particles are emitted at the altitude of clouds that can exert a strong radiative influence. This subject is examined by Kärcher and Spichtinger, this volume, and Denman et al., 2007, and is thus not reviewed here.

5. Global Climate Model Estimates of the Total Anthropogenic Aerosol Effect

The total anthropogenic aerosol effect as defined here consists of the direct effect, semi-direct effect, indirect cloud albedo effect and cloud lifetime effect for warm clouds. The total anthropogenic aerosol effect is obtained as the difference between a multi-year simulation with present-day aerosol emissions and a simulation representative for preindustrial conditions, in which anthropogenic emissions are turned off. It should be noted that the representation of the cloud lifetime effect in global climate models (GCMs) is essentially one of changing the autoconversion of cloud water to rainwater.

The radiative forcing resulting from the indirect cloud albedo effect due to anthropogenic aerosols is estimated from global models as –0.7 W m^{-2} with a 90% confidence range of –0.3 to –1.8 W m^{-2} (Forster et al., 2007). Feedbacks due to the cloud lifetime effect, semi-direct effect or aerosol-ice cloud effects can either enhance or reduce the cloud albedo effect. Climate models estimate the total aerosol effect (direct plus indirect effects) on the top-of-the-atmosphere net radiation since pre-industrial times to be –1.2 W m^{-2} with a range of –0.2 to –2.3 W m^{-2} (Figure 6 and Denman et al., 2007). The range of the total aerosol effect from different models cannot easily be compared to the range of the indirect cloud albedo effect alone because different model simulations entered these different compilations.

All models agree that the total aerosol effect is larger over the Northern Hemisphere than over the Southern Hemisphere (Figure 6), consistent with emissions of anthropogenic aerosols and precursor gases being much greater in the Northern Hemisphere. However, such an effect has not been seen in satellite data (Han et al., 1998; Schwartz, 1988) suggesting that dynamical influences on the liquid water path mask such an effect. The values of the Northern Hemisphere total aerosol effect vary between –0.5 and –3.6 W m^{-2} and on the Southern Hemisphere between slightly positive to –1.1 W m^{-2} with an average Southern Hemisphere to Northern Hemisphere ratio of 0.3. Estimates of the ocean/land partitioning of the total aerosol effect vary from 0.03 to 1.8 with an average value of 0.7. While the combined ECHAM4 GCM+POLDER satellite estimate suggests that the total aerosol effect should be larger over oceans, combined estimates of the LMD and ECHAM4 GCMs with MODIS satellite data reach the opposite conclusion. The average total aerosol effect over the ocean of –1 W m^{-2} agrees with estimates between –1 to –1.6 W m^{-2} from AVHRR/POLDER (Denman et al., 2007).

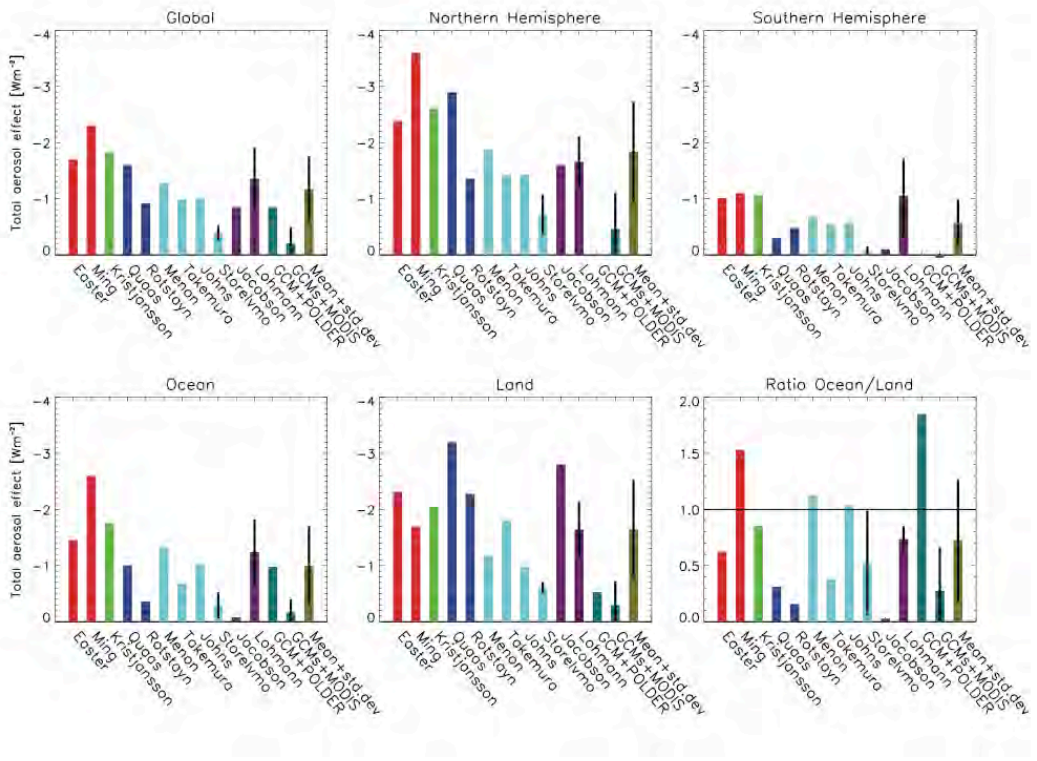

Figure 6: Total anthropogenic aerosol effect (direct, semi-direct and indirect cloud albedo and lifetime effects) in 12 global climate models and two determinations from satellite observations in global mean, over the Northern and Southern Hemisphere, over oceans and over land, and the ratio over oceans/land. Anthropogenic aerosol effect is defined as the change in net radiation at TOA from pre-industrial times to the present day resulting from anthropogenic emissions of aerosols and aerosol precursors. Colors of bars for models denote different anthropogenic species whose forcings were examined and the cloud types affected; all are for water clouds except as indicated. Red bars denote results for models examining the effect of anthropogenic sulphate; green, anthropogenic sulphate and black carbon; blue, anthropogenic sulphate and organic carbon; turquoise, anthropogenic sulphate and black and organic carbon; dark purple, anthropogenic sulphate and black and organic carbon effects on water and ice clouds. Teal bars denote a combination of GCM and satellite results, and olive bars denote the mean and standard deviation from all simulations. Vertical black lines denote ±1 standard deviation in cases of multiple simulations and/or results. See Denman et al. (2007) for details.

GCM estimates of the total aerosol effect are generally larger than estimated from inverse models which constrain the indirect aerosol effect to be between –0.1 and –1.7 W m^{-2} (Anderson et al., 2003; Hegerl et al, 2007). The estimated total anthropogenic aerosol effect is now lower than in the Third Assessment report of IPCC mainly because of improvements in cloud parametrizations, but large uncertainties remain.

The influence of aerosols on evapotranspiration and precipitation is also quite uncertain, with model results for the change in global average precipitation ranging from almost no change to a decrease of 0.13 mm day^{-1} (5 cm yr^{-1}), with much greater changes locally. Decreases in precipitation are larger when the atmospheric GCMs are coupled to mixed-layer ocean models where the sea surface temperature and, hence, the evaporation from the ocean is also allowed to vary, than in models in which the sea surface temperature is held constant (Denman et al., 2007). The decrease in evapotranspiration results mainly from decreases in solar radiation at

the surface due to the increased aerosol optical depth and the optically thicker clouds. The decrease in solar radiation at the surface is then partly balanced by a decreased latent heat flux, which results in a reduced global mean precipitation rate (e.g., Liepert et al., 2004).

6. Cloud feedbacks in climate models and their influence on climate sensitivity

The energy balance model of Earth's climate system is a useful means of assessing the influences of particular processes on global mean surface temperature (T) and on comparing these influences across different climate models. Within this energy balance framework the time-dependent change in heat content of the climate system ΔQ is related to the radiative forcing ΔF and the change in T ΔT as

$$\frac{d}{dt}(\Delta Q) = \Delta F - \frac{1}{\lambda}\Delta T \tag{3}$$

where λ is the equilibrium climate sensitivity, as is readily seen by considering a system in a new equilibrium in response to a forcing ΔF, for which $d(\Delta Q)/dt = 0$ and hence

$$\lambda = \frac{\Delta T_{\mathrm{eq}}}{\Delta F} \tag{4}$$

where ΔT_{eq} is the temperature difference between two equilibrium states. At present climate sensitivity is not well constrained either in climate models or in empirical analyses. According to the IPCC 2007 assessment report (IPCC, 2007), the likely range of global equilibrium temperature increase for doubling of CO_2 $\Delta T_{2\times}$ is between 2.0 and 4.5 K, with values below 1.5 K considered very unlikely. This sensitivity range is consonant with values exhibited by current GCMs, Figure 7. As a doubling of CO_2 causes direct radiative forcing of about 3.7 W m^{-2} (Forster et al, 2007), the range of 2.0 to 4.5 K for doubling of CO_2 corresponds to climate sensitivity between 0.54 and 1.22 K/(W m^{-2}). As pointed out by Roe and Baker (2007), the upper range of the climate sensitivity is relatively insensitive to decreases in uncertainties associated with the underlying climate processes. In this context it is notable that some recent experiments with cloud resolving models embedded within GCMs (Miura et al., 2005; Wyant et al., 2006) suggest lower climate sensitivity, with values of 0.44 and 0.41 K/(W m^{-2}), respectively ($\Delta T_{2\times}$ 1.6 and 1.5 K). These findings point to the strong influence of the treatment of clouds on modeled climate sensitivity.

Cloud feedback is the response of cloud radiative forcing (CRF) to a change in global temperature. Key questions involve the nature and extent of CRF changes in a greenhouse-warmed world: As the climate warms will longwave CRF increase (positive feedback) or decrease (negative feedback); likewise will shortwave CRF increase (negative feedback) or decrease (positive feedback)? Current climate models produce a wide variety of cloud feedbacks ranging from weakly negative to strongly positive, depending on the relative magnitude of different cloud feedback mechanisms (Bony et al. 2006; Webb et al. 2006). As seen in Figure 7, much of the model-to-model difference in climate sensitivity is due to differences in cloud feedback.

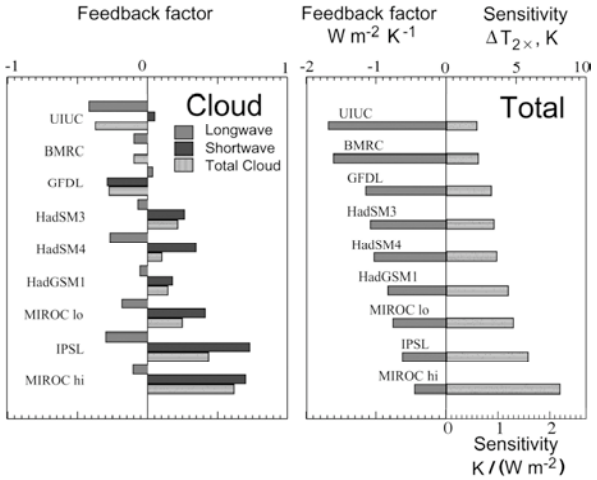

Figure 7. Influence of cloud feedback on total feedback and sensitivity of current GCMs. Left panel shows total cloud feedback and long- and shortwave components in nine current climate models. Right panel shows total feedback factor and sensitivity expressed as the inverse of the total feedback factor, in units of K /(W m^{-2}), and as the equilibrium increase in global mean surface temperature that would result from a doubling of CO_2, ΔT_{2x}, evaluated as –3.7 W m^{-2} upon the total feedback factor. Modified from Webb et al. 2006.

CRF depends largely on the spatial and temporal distribution of clouds and their radiative properties, which are determined by microphysical characteristics such as size distribution of droplets and ice crystals. These cloud properties are controlled by such cloud-forming processes as cooling by rising air motion or radiative cooling and by such cloud-dissipating processes as precipitation, sinking motion, and mixing with dry air. Cloud feedbacks thus involve changes in the spatial distribution of clouds and their microphysical properties resulting from changes in processes that form and dissipate clouds. Uncertainty in cloud feedback arises from uncertainty in how cloud microphysical properties and cloud horizontal and vertical distributions respond to changes in controlling variables as the climate changes. In contrast to CRF, cloud feedback cannot be measured directly and hence can be determined only from GCM simulations. Consequently confidence in estimates of cloud feedback can be assessed only by using observations to evaluate simulations of cloud microphysical and radiative properties, cloud distribution, and radiative forcing in a variety of conditions that span the range expected under climate change. Atmospheric process research on cloud feedbacks therefore focuses on how these cloud features depend on processes that form and dissipate clouds under a variety of conditions.

Closely related to cloud feedback is water-vapor feedback. Water vapor is a greenhouse gas (GHG) and indeed the most important GHG in Earth's atmosphere. Although all climate models show an increase in the amount of atmospheric water vapor with rising global mean surface temperature, the amount and spatial distribution of the resultant radiative forcing differ considerably among models. This water-vapor feedback is strongly interactive with cloud feedback because water vapor is the source of condensed-phase water in clouds, and clouds remove water from the atmosphere when they precipitate. Clouds also influence evapotranspiration, which supplies water vapor to the atmosphere.

7. Conclusions and outlook

In summary clouds play a key role in determining planetary albedo and the absorption of solar radiation that must be accurately represented in climate models if these models are to be used with confidence in projections of future climate change. Indeed much of the variation in

climate sensitivity among present climate models is attributable to differences in the treatment of clouds, as manifested by model-to-model variation in the cloud feedback factor. It is recognized as well that cloud microphysical processes exert strong influences on cloud dynamics and the influences of clouds on short- and longwave radiation that must be understood and accurately represented in climate models. There is as well a strong coupling between aerosols and clouds with resultant large influences on climate and climate change. An increased loading of atmospheric aerosol changes cloud microphysical properties with resultant influences on short- and longwave radiation and on precipitation development, affecting changes in the locus and intensity of precipitation. The best understood influences of aerosols on clouds are a reduction of the amount of solar radiation absorbed by the Earth-atmosphere system, as quantified by net shortwave radiation at the top-of-the-atmosphere and a similar decrease in shortwave radiation reaching the surface. The negative radiative forcing of anthropogenic aerosols competes with greenhouse gas warming as a forcing of climate change and also in determining changes in evaporation and precipitation of water. Although much has been learned about these effects in recent years, they are not well understood at present, and as a consequence are at best only incompletely represented in climate models. None of the transient climate model simulations that have been conducted thus far account for all the known aerosol-cloud interactions, so that the net effects of aerosols on clouds and climate deduced from global climate models cannot be considered conclusive. For these reasons as well the cloud feedback and sensitivity of Earth's climate system remain highly uncertain.

The key areas for future development in terms of aerosols and clouds in GCMs are twofold. Treatments of clouds by themselves need to be improved in all aspects if models are to accurately represent cloud feedbacks in a greenhouse warmed world. A good representation of cloud dynamics including entrainment is important especially for the representation of convective clouds and boundary layer clouds. Cloud microphysical processes are important for the conversion of cloud particles into precipitation-size particles. At the same time, treatment of aerosol-cloud-interactions needs to be improved if aerosol radiative forcing is to be accurately quantified.

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